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## PATENT COOPERATION TREATY

PCT

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## INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference CDK 1693	<b>FOR FURTHER ACTION</b> See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)	
International application No. PCT/GB00/02448	International filing date (day/month/year) 22/06/2000	Priority date (day/month/year) 24/06/1999
International Patent Classification (IPC) or national classification and IPC C11D17/00		
Applicant RHODIA CONSUMER SPECIALTIES LIMITED et al.		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.



2. This REPORT consists of a total of 6 sheets, including this cover sheet.

- ☐ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

These annexes consist of a total of sheets.

3. This report contains indications relating to the following items:

- I ☒ Basis of the report
- II ☐ Priority
- III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- IV ☐ Lack of unity of invention
- V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- VI ☐ Certain documents cited
- VII ☒ Certain defects in the international application
- VIII ☒ Certain observations on the international application

Date of submission of the demand  20/2/2000	Date of completion of this report  10.08.2001
Name and mailing address of the international preliminary examining authority:  European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Authorized officer  Ruckebusch, V  Telephone No. +49 89 2399 8493 

# INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/GB00/02448

## I. Basis of the report

1. With regard to the **elements** of the international application (*Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17)*):

**Description, pages:**

1-17 as originally filed

**Claims, No.:**

1-9 as originally filed

2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language: , which is:

- ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).
- ☐ the language of publication of the international application (under Rule 48.3(b)).
- ☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

- ☐ contained in the international application in written form.
- ☐ filed together with the international application in computer readable form.
- ☐ furnished subsequently to this Authority in written form.
- ☐ furnished subsequently to this Authority in computer readable form.
- ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
- ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

- ☐ the description, pages:
- ☐ the claims, Nos.:
- ☐ the drawings, sheets:

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

# INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/GB00/02448

*(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)*

6. Additional observations, if necessary:

## V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

### 1. Statement

Novelty (N)	Yes:	Claims	1-9
	No:	Claims	
Inventive step (IS)	Yes:	Claims	
	No:	Claims	1-9
Industrial applicability (IA)	Yes:	Claims	1-9
	No:	Claims	

### 2. Citations and explanations **see separate sheet**

## VII. Certain defects in the international application

The following defects in the form or contents of the international application have been noted:  
**see separate sheet**

## VIII. Certain observations on the international application

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:  
**see separate sheet**

**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT - SEPARATE SHEET**

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International application No. PCT/GB00/02448

- i. Reference is made to the following documents:

D1: WO 91 08281 A (UNILEVER ) 13 June 1991 (1991-06-13)

D2: US-A-4 931 195 (Cited by the Examining Division)

**Re Item V**

Reasoned statement under Rule 66.2(a)(ii) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

**v. Inventive Step (Article 33(3) PCT)**

- v.i All the examples of the present application comprise a deflocculating polymer and some cellulose derivative.

The Applicant observed that compositions, devoid of Accusol 842, but comprising greater amounts (though not quantified) of Laponite RD were satisfactory (Example 2).

The Applicant thus considers that the substitution of a part of said clay by a lower amount of Accusol 842 amounts to an unexpected achievement which could amount to an inventive step.

- v.ii Structured detergent compositions comprising such a modified clay are known from the art, see D2 (being concerned with stable liquid detergent compositions).
- v.iii It may be noted that D1 discloses many structured compositions (see Example 1 A to D; Example 2 D and H) and teaches that structured, deflocculated compositions may be further stabilised with a cellulosic derivative (identified in the present application as soil release polymer; that effect is nevertheless compatible with an improved stabilisation).
- v.iv Acusol 842 is acknowledged by the producer as a viscosity modifier and stabiliser for detergent compositions.

**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT - SEPARATE SHEET**

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International application No. PCT/GB00/02448

- v.v** It appears thus that the compositions of Examples 1 and 2 of the present application are nothing more than a juxtaposition of features known from the art (viz. the two documents referred to above and the property of Acusol 842), functioning their own and known way !

The Applicant did not solve any problem which could not have been solved by the combination of the two documents referred to ; he did not establish that the components of his compositions together act in synergy; the subject-matter of the present application thus lacks an inventive step.

His considerations about the respective prices of Acusol and modified clays may be relevant for a detergent composition producer; but they are not of a technical (in the sense "solution of technical problem") nature.

**Re Item VII**

Certain defects in the international application

- vii.** The Applicant did not gave any reason, why the relevant background art disclosed in the document D1 is not mentioned in the description, nor are these documents identified therein, contrary to the requirements of Rule 5.1(a)(ii) PCT.

**Re Item VIII**

Certain observations on the international application

- viii.** The Examining Division cannot see how Acusol 842 can be defined as a "polymer having a hydrophilic backbone"; the drafting of claim 1 is misleading and does not reflect the investigations of the Applicant.
- 

The Applicant investigated Acusol 842 only; his conclusions (whatever they may be) about different polymers are speculative, because not established.

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Incidentally, the subject-matter of Claims 1 and 9 is not clear because defined in terms of the result to be achieved ("structured", "capable of suspending solids", "to form a flocculated, dispersed...system" and "inhibit the flocculation of said system") which terms merely amount to a statement of the underlying problem. The technical features necessary for achieving this result are not specified/identified.

(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization  
International Bureau



(43) International Publication Date  
4 January 2001 (04.01.2001)

PCT

(10) International Publication Number  
**WO 01/00779 A1**

- (51) International Patent Classification<sup>7</sup>: C11D 17/00, 3/37, 3/12
- (74) Agent: BARKER BRETTELL; 138 Hagley Road, Edgbaston, Birmingham B16 9PW (GB).
- (21) International Application Number: PCT/GB00/02448
- (22) International Filing Date: 22 June 2000 (22.06.2000)
- (25) Filing Language: English
- (26) Publication Language: English
- (30) Priority Data: 9914671.4 ✓ 24 June 1999 (24.06.1999) GB
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- (81) Designated States (national): AE, AG, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, BZ, CA, CH, CN, CR, CU, CZ, DE, DK, DM, DZ, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, MZ, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, TZ, UA, UG, US, UZ, VN, YU, ZA, ZW.
- (84) Designated States (regional): ARIPO patent (GH, GM, KE, LS, MW, MZ, SD, SL, SZ, TZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).
- Published:**  
— With international search report.
- For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

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WO 01/00779 A1

(54) Title: STRUCTURED SURFACTANT SYSTEMS

(57) Abstract: Structured surfactant systems contain a deflocculant and, in addition, an auxiliary stabiliser to inhibit temperature instability. The auxiliary stabiliser comprises a substantially non cross-linked water soluble copolymer having a hydrophilic backbone and sufficient C<sub>2-6</sub> hydrophobic organic side chains to permit entanglement of the polymer chains.



## STRUCTURED SURFACTANT SYSTEMS

The present invention relates to the formulation of structured surfactant suspending systems. It is particularly relevant to the formulation of laundry detergents especially those used for industrial and institutional cleaning.

### STRUCTURED SURFACTANT

Suspending solids in liquids presents a problem. If the solids differ in density from the liquid they will tend either to sediment or float. Increasing the viscosity of the liquid can retard, but not prevent such separation, and high viscosities are generally undesirable. Colloidal systems, in which the suspended particles are sufficiently small to experience Brownian motion, e.g. less than 1 micron, may be kinetically stable. However the difficulty or undesirability of comminuting some solids to such sizes, and the impossibility of maintaining many of them at this level in the face of crystal growth or agglomeration, limits the use of colloidal suspensions.

Adjusting the density of one phase to match that of the other is usually impracticable. Moreover such systems are almost always temperature-unstable due to differential rates of thermal expansion.

One method of suspension which permits even relatively large particles to be stably suspended is structured surfactant. The term covers systems in which a surfactant mesophase, usually a lamellar or G-phase, alone or more usually interspersed with an aqueous phase, provides a yield stress which is sufficient, when the system is at rest, to immobilise any suspended particles, but which is sufficiently low to allow the system to be poured like a normal liquid. Such systems may display very low apparent viscosities when stirred, pumped or poured and yet be capable of maintaining particles, sometimes of millimetre or larger size, indefinitely in suspension.

Three main types of suspending system have been employed in practice, all involving a G-phase, in which bilayers of surfactant are arranged with the hydrophobic part of the molecule on the interior and the hydrophilic part on the exterior of the bilayer (or vice versa). The bilayers lie side by side, e.g. in a parallel or concentric configuration, sometimes separated by aqueous layers. G-phases (also known as  $L_{\alpha}$  phases) can usually be identified by their characteristic textures under the polarising microscope and/or by x-ray diffraction, which is often able to detect evidence of lamellar symmetry. Such evidence may comprise first, second and sometimes third order peaks with d-spacing ( $\frac{2\pi}{Q}$  where  $Q$  is the momentum transfer vector) in a simple integral ratio 1:2:3. Other types of symmetry give different ratios, usually non integral.

Most surfactants form a G-phase either at ambient or at some higher temperature when mixed with water in certain specific proportions. However such G-phases cannot usually be used as structured suspending systems. Useful quantities of solid render them unpourable and smaller amounts tend to sediment.

The main types of structured system used in practice are based on dispersed lamellar, spherulitic and attenuated lamellar phases. Dispersed lamellar phases are two phase systems in which the surfactant bilayers are arranged as parallel plates to form domains of G-phases which are interspersed with an aqueous phase to form an opaque gel-like system. They are described in EP O 086 614.

Spherulitic phases comprise well defined spheroidal bodies, usually referred to in the art as spherulites, in which surfactant bilayers are arranged as concentric shells. The spherulites usually have a diameter in the range 0.1 to 15 microns and are dispersed in an aqueous phase in the manner of a classical emulsion, but interacting to form a structured system. Spherulitic systems are described in more detail in EP O 151 884.

Many structured surfactant systems are intermediate between dispersed lamellar and spherulitic, involving both types of structure. Usually systems having a more Spherulitic character are preferred because they tend to have lower viscosity. A variant on the spherulitic system comprises prolate or rod shaped bodies sometimes referred to as batonets.

A third type of structured surfactant system comprises an expanded G-phase. It differs from the other two types of structure system in being essentially a single phase, and from conventional G-phase in having a wider d-spacing. Conventional G-phases have a d-spacing of about 5 to 7 nanometers. Attempts to suspend solids in such phases results in stiff pastes which are either non-pourable, unstable or both. Expanded G-phases with d-spacing between 8 and 20, e.g. 10 to 15 nanometers, form when the electrolyte is added to aqueous surfactants at concentrations just below those required to form a normal G-phase, particularly to surfactants in the M phase. The M phase comprises surfactant molecules arranged to form cylindrical rods of indefinite length. It exhibits hexagonal symmetry and a distinctive texture under the polarising microscope. Typical M phases have so high a viscosity that they appear to be curdy solids. M phases near the lower concentration limit (the  $L_1$ /M phase boundary) may be pourable but have a very high viscosity and often a mucous-like appearance. Such systems tend to form expanded G-phases particularly readily on addition of sufficient electrolyte.

Expanded G-phases are described in more detail in EP O 530 708. In the absence of suspended matter they are translucent, unlike dispersed lamellar or spherulitic phases which are necessarily opaque. They are optically anisotropic and have shear dependent viscosity. ~~In this they differ from  $L_1$  phases which are micellar solutions~~ and which include microemulsions.  $L_1$  phases are clear, optically isotropic and substantially Newtonian. They are unstructured and cannot suspend solids. Some  $L_1$  phases exhibit small angle x-ray diffraction spectra which show evidence of hexagonal symmetry. Such phases usually have concentrations near the  $L_1$ /M phase boundary and may form expanded G-phases on addition of electrolyte.

Most structured surfactant systems require the presence of electrolyte as well as surfactant and water in order to form structured systems capable of suspending solids. However certain relatively hydrophobic surfactants such as isopropylamine alkyl benzene sulphonate can form spherulites in water in the absence of electrolyte. Such surfactants are capable of suspending solids in the absence of electrolyte as described in EP O 414 549.

### **THE PROBLEM**

A problem with the two phase structured surfactant systems, and especially spherulitic systems, is flocculation of the dispersed surfactant structures. This tends to occur at high surfactant and/or high electrolyte concentration. It can have the effect of making the composition very viscous and/or unstable with the dispersed surfactant separating from the aqueous phase.

### **THE PRIOR ART**

Certain amphiphilic polymers have been found to act as deflocculants of structured surfactants. One type of deflocculant polymer exhibits cteniform (comb-shaped) architecture with a hydrophilic backbone and hydrophobic side chains or vice versa. A typical example is a random copolymer of acrylic acid and a fatty alkyl acrylate. Cteniform deflocculants have been described in a large number of patents, for example WO-A-9106622.

A more effective type of deflocculant has surfactant rather than cteniform architecture. With a hydrophilic polymer group attached at one end to a hydrophobic group. Such deflocculants are typically telomers formed by telomerising a hydrophilic monomer with a hydrophobic telogen. Examples of surfactant deflocculants include alkyl thiol polyacrylates and alkyl polyglycosides. Surfactant deflocculants are described in more details in EP O 623 670.

A disadvantage of both surfactant and cteniform deflocculants is that the concentration required to deflocculate to the extent required for optimum viscosity is critical within fairly narrow limits and varies with temperature. Either too much or too little deflocculant causes instability and/or excessive viscosity. As a result the deflocculated systems tend to separate or congeal if the temperature varies significantly. In particular, separation of a clear bottom layer is often observed on storage.

One approach to the problem of temperature stability has been to add highly cross linked polyacrylates (see US 5 602 092). These, however, are difficult to disperse in the structured liquid.

Structured surfactants have been applied to the problems of suspending: water insoluble or sparingly soluble builders in laundry detergent; antifoams and enzymes in laundry detergents and other surfactant systems; abrasives in hard surface cleaners; pesticides and oils in agrochemical preparations; rock cuttings in drilling muds; dyestuffs in dyebath concentrates and printing inks; talcs, oils and other cosmetic ingredients in personal care formulations.

### **THE SOLUTION**

We have now discovered that a substantially non-cross linked polymer having a hydrophilic backbone and sufficient short (e.g. C<sub>1 to 5</sub>) side chains to enhance physical entanglement of the polymer molecules has the ability to stabilise deflocculated structured surfactant systems, but is more soluble and easier to disperse than the cross linked polymers used hitherto.

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## **THE INVENTION**

Our invention provides a structured surfactant composition capable of suspending solids which comprises surfactant water and, if required, electrolyte in relative proportions adapted to form a flocculated, dispersed lamellar and/or spherulitic structured surfactant system, and, in addition, sufficient of a deflocculant to inhibit the flocculation of said system characterised in that said composition comprises an effective amount of an auxiliary stabiliser which is a substantially non-cross linked, water-soluble copolymer having a hydrophilic backbone and sufficient C<sub>2-6</sub> hydrophobic organic side chains to permit entanglement of the polymer chain. For convenience as used herein "side chain" includes cyclic side chains such as a benzene, cyclohexane or cyclopentane ring, attached to the hydrophilic backbone.

## **THE AUXILIARY STABILISER**

The auxiliary stabiliser may be a copolymer of (a) one or more unsaturated carboxylic or dicarboxylic acids having from 3 to 6 carbon atoms and/or vinyl alcohol with (b) styrene and/or a C<sub>1 to 5</sub> alkyl ester of a C<sub>3 to 6</sub> unsaturated carboxylic or dicarboxylic acid and/or a vinyl alcohol ester of a C<sub>2 to 6</sub> carboxylic acid wherein the mole ratio of (a):(b) is from 0.01 to 10, preferably more than 0.1, especially more than 0.2, e.g. more than 0.5, but preferably less than 8, especially less than 5, e.g. less than 2. The polymer is substantially non-cross linked by which is meant that it comprises less than 0.05% cross linking and/or insufficient cross linking to provide pseudo plastic behaviour. Preferably the auxiliary stabiliser gives the formulation a Sisko Index of 0.1 to 0.4, e.g. 0.2 to 3.

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The polymer is preferably present in an amount of from 0.01 to 10% by weight of the composition e.g. 0.05 to 5%, especially 0.1 to 2%, e.g. 0.5 to 1.5% by weight.

The polymer may preferably be a copolymer of acrylic acid with for example ethyl, propyl or butyl acrylate and/or styrene. Alternatively, a copolymer comprising vinyl alcohol and vinyl acetate propionate or butyrate residues could be used. Other

monomers include maleic acid, fumaric acid, citraconic acid, aconitic acid, itaconic acid, crotonic acid, isocrotonic acid, angelic acid and tiglic acid and their esters or partial esters. The effectiveness of the auxiliary stabiliser may be enhanced by the presence of a clay such as bentonite. The latter, however, is not normally sufficient to stabilise the composition in the absence of the polymer. Particularly preferred are water dispersible synthetic layered silicates such as that sold by Laporte under its Registered Trade Mark "LAPONITE".

It has been found that clays and, in particular, synthetic layered silicates are synergistic with the copolymeric auxiliary stabilisers. This provides a further aspect of the invention. The clay is generally used in amounts between 0.01 and 10% by weight of the composition. Typically using clays such as bentonite, proportions of from 0.1 to 8% may be used, more usually 0.5 to 5%, e.g. 1 to 3%. However, synthetic layered silicates are effective in substantially lower concentrations e.g. 0.01 to 2%, e.g. 0.05 to 1%, especially 0.1 to 0.5% by weight. Use of the clay or silicate permits the proportion of copolymer to be substantially reduced, e.g. 0.01 to 0.5% by weight based on the weight of the composition.

The invention therefore provides, according to a further embodiment an auxiliary stabiliser for deflocculated structured surfactants which comprises from 10 to 90% by weight of a copolymer having a hydrophilic backbone and  $C_{2 \text{ to } 6}$  hydrophobic side chains, with from 90 to 10% by weight of a natural or synthetic clay.

### **DEFLOCCULANT**

The deflocculant may be a surfactant deflocculant including any of those referred to in EP O 623 670. Particularly preferred are the alkyl thiol polycarboxylate telomers such as a  $C_{8-20}$  alkyl thiol polyacrylate or polymaleate and alkyl polyglycosides such as  $C_{8-20}$  alkyl polyglucoside e.g. having a D.P. greater than 1.2, preferably greater than 1.5. Alternatively the stabiliser may be a cteniform polymer of the type described in WO-A-9106622. The deflocculant is typically present in the amount required to deflocculate the system. This depends on the nature of the surfactant and of the

deflocculant but is typically in the range 1 to 5% although higher concentrations may be required in some cases.

## **SURFACTANT**

Compositions according to the present invention generally contain at least sufficient surfactant to form a structured system. For some surfactants this may be as low as 2% by weight, but more usually requires at least 3%, especially at least 4%, typically more than 5% by weight of surfactant.

Detergent compositions of the present invention preferably contain at least 10% by weight of total surfactant (including the deflocculant, where the latter is a surfactant) based on the total weight of the composition. Most preferably the total surfactant is at least 20%, especially more than 25%, e.g. more than 30% by weight of the composition. It is unlikely in practice that the surfactant concentration will exceed 80% based on the weight of the composition and is usually less than 70% especially less than 60%, typically less than 50% e.g. less than 40% by weight of the composition.

The amount of surfactant present in the composition is preferably greater than the minimum which is able, in the presence of a sufficient quantity of surfactant-desolubilising electrolyte, to form a stable, solids-suspending structured surfactant system.

The surfactant may comprise anionic, cationic, non-ionic, amphoteric semi polar and/or zwitterionic species or mixtures thereof.

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Anionic surfactant may comprise a C<sub>10-20</sub> alkyl benzene sulphonate or an alkyl ether sulphate which is preferably the product obtained by ethoxylating a natural fatty or synthetic C<sub>10-20</sub> e.g. a C<sub>12-14</sub> alcohol with from 1 to 20, preferably 2 to 10 e.g. 3 to 4 ethyleneoxy groups. optionally stripping any unreacted alcohol, reacting the ethoxylated product with a sulphating agent and neutralising the resulting alkyl ether



sulphuric acid with a base. The term also includes alkyl glyceryl sulphates, and random or block copolymerised alkyl ethoxy/propoxy sulphates.

The anionic surfactant may also comprise, for example,  $C_{10-20}$  e.g.  $C_{12-18}$  alkyl sulphate.

The surfactant may comprise a  $C_{8-20}$  e.g.  $C_{10-18}$  aliphatic soap. The soap may be saturated or unsaturated, straight or branched chain.

Preferred examples include dodecanoates, myristates, stearates, oleates, linoleates, linolenates and palmitates and coconut and tallow soaps. Where foam control is a significant factor we particularly prefer to include soaps e.g. ethanolamine soaps and especially monoethanolamine soaps, which have been found to give particularly good cold storage and laundering properties.

The surfactant may include other anionic surfactants, such as olefin sulphonates, paraffin sulphonates, taurides, isethionates, ether sulphonates, ether carboxylates, aliphatic ester sulphonates, e.g. alkyl glyceryl sulphonates, sulphosuccinates or sulphosuccinamates. Preferably the other anionic surfactants are present in total proportion of less than 45% by weight, based on the total weight of surfactants, more preferably less than 40%, most preferably less than 30%, e.g. less than 20%.

The cation of any anionic surfactant is typically sodium but may alternatively be potassium, lithium, calcium, magnesium, ammonium, or an alkylammonium having up to 6 aliphatic carbon atoms including isopropylammonium, monoethanolammonium, diethanolammonium, and triethanolammonium. Mixtures of the above cations may be used.

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The surfactant preferably contains one, or preferably more, non-ionic surfactant. These preferably comprise alkoxylated C<sub>8-20</sub> preferably C<sub>12-18</sub> alcohols. The alkoxylates may be ethoxylates, propoxylates or mixed ethoxylated/propoxylated alcohols. Particularly preferred are ethoxylates with 2 to 20 especially 2.5 to 15 ethyleneoxy groups.

The alcohol may be fatty alcohol or synthetic e.g. branched chain alcohol. Preferably the non-ionic component has an HLB of from 6 to 16.5, especially from 7 to 16, e.g. 8 to 15.5. We particularly prefer mixtures of two or more non-ionic surfactants having a weighted mean HLB in accordance with the above values.

Other ethoxylates and/or propoxylated non-ionic surfactants which may be present include C<sub>6-16</sub> alkylphenol alkoxylates, alkoxylated fatty acids, alkoxylated amines, alkoxylated alkanolamides and alkoxylated alkyl sorbitan and/or glyceryl esters.

Other non-ionic surfactants which may be present include amine oxides, fatty alkanolamides such as coconut monoethanolamide, and coconut diethanolamide and alkylaminoethyl fructosides and glucosides.

The proportion by weight of non-ionic surfactant is preferably at least 2% and usually more than 10%, more typically more than 20%, e.g. 30 to 75%, especially 40 to 60% based on the total weight of surfactant. However compositions wherein the non-ionic surfactant is from 75 to 100% of the total weight of the surfactant are included and may be preferred for some applications.

The surfactant may be, or may comprise major or minor amounts of, amphoteric and/or cationic surfactants, for example betaines, sulphobetaines, amidobetaines, imidazolines, amidoamines, quaternary ammonium surfactants and cationic fabric conditioners having two long chain alkyl groups, such as tallow groups. Examples of fabric conditioners which may be deflocculated according to our invention include ditallowyl dimethyl ammonium salts, ditallowyl methyl benzyl ammonium salts, ditallowyl imidazolines, ditallowyl amidoamines and quaternised ditallowyl

imidazolines and amidoamines. The anion of the fabric conditioner may for instance be or may comprise methosulphate, chloride, sulphate, acetate, lactate, tartrate, citrate or formate. We prefer that the compositions of our invention do not contain substantial amounts of both anionic and cationic surfactants.

We particularly prefer that the surfactant consists essentially of a mixture of a non-ionic ethoxylate with an amine oxide or, preferably an amphoteric surfactant. Such mixtures exhibit strong synergistic soil removal. The mixture may desirably contain from 1:10 to 10:1 weight ratio of non-ionic to amphoteric e.g. 1:5 to 5:1.

### **SUSPENDED SOLIDS**

A major advantage of the preferred compositions of the invention is their ability to suspend solid particles to provide non-sedimenting pourable suspension. Optionally the composition may contain up to, for example, 80% by weight, based on the weight of the composition, of suspended solids, more usually up to 30 e.g. 10 to 25%. The amount will depend on the nature and intended use of the composition. For example in detergent compositions it is often desired to include insoluble builder such as zeolite or sparingly soluble builders such as sodium tripolyphosphate which may be suspended in the structured surfactant medium.

The surfactant systems according to our invention may also be used to suspend abrasives such as talc, silica, calcite or coarse zeolite to give hard surface cleaners; or pesticides, to provide water dispersible, pourable compositions containing water insoluble pesticides, without the hazards of toxic dust or environmentally harmful solvents. They are useful in providing suspensions of pigments, dyes, pharmaceuticals, biocides, or as drilling muds, containing suspended shale and/or weighting agents such as sodium chloride, calcite, barite, galena or haematite.

They may be used to suspend exfoliants including talc, clays, polymer beads, sawdust, silica, seeds, ground nutshells or dicalcium phosphate, pearlisers such as mica, glycerol mono- or di-stearate or ethylene glycol mono- or di-stearate, natural oils,

including mineral and glyceride oils such as coconut, evening primrose, groundnut, meadow foam, apricot kernel, avocado, peach kernel or jojoba oils, essential oils, synthetic oils such as silicone oils, vitamins, anti-dandruff agents such as zinc omadine, and selenium disulphide, proteins, emollients such as lanolin or isopropylmyristate, waxes and sunscreens such as titanium dioxide and zinc oxide.

### **BUILDERS**

We prefer that detergent compositions of our invention contain dissolved builder and/or suspended particles of solid builder, to provide a fully built liquid detergent. "Builder" is used herein to mean a compound which assists the washing action of a surfactant by ameliorating the effects of dissolved calcium and/or magnesium. Generally builders also help maintain the alkalinity of wash liquor. Typical builders include sequestrants and complexants such as sodium tripolyphosphate, potassium pyrophosphate, trisodium phosphate, sodium ethylene diamine tetracetate, sodium citrate or sodium nitrilo-triacetate, ion exchangers such as zeolites and precipitants such as sodium or potassium carbonate and such other alkalis as sodium silicate. Said stabiliser also contributes to the total builder. The preferred builders are zeolite and sodium tripolyphosphate. The builder may typically be present in concentrations up to 50% by weight of the composition e.g. 15 to 30%.

### **pH**

The pH of a composition for laundry use is preferably alkaline, as measure after dilution with water to give a solution containing 1% by weight of the composition, e.g. 7 to 12, more preferably 8 to 12, most preferably 9 to 11.

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## **HYDROTROPES**

Compositions of our invention may optionally contain small amounts of hydrotropes such as sodium xylene sulphonate, sodium toluene sulphonate or sodium cumene sulphonate, e.g. in concentrations up to 5% by weight based on the total weight of the composition, preferably not more than 2%, e.g. 0.1 to 1%. Hydrotropes tend to break surfactant structure and it is therefore important not to use excessive amounts. They are primarily useful for lowering the viscosity of the formulation, but too much may render the formulation unstable.

## **SOLVENTS**

The compositions may contain solvents, in addition to water. However, like hydrotropes, solvents tend to break surfactant structure. Moreover, again like hydrotropes, they add to the cost of the formulation without substantially improving the washing performance. They are moreover undesirable on environmental grounds and the invention is of particular value in providing solvent-free compositions. We therefore prefer that they contain less than 6%, more preferably less than 5%, most preferably less than 3%, especially less than 2%, more especially less than 1%, e.g. less than 0.5% by weight of solvents such as water miscible alcohols or glycols, based on the total weight of the composition. We prefer that the composition should essentially be solvent-free, although small amounts of glycerol and propylene glycol are sometimes desired. Concentrations of up to about 3% by weight, e.g. 1 to 2% by weight of ethanol are sometimes required to enhance perfume. Such concentrations can often be tolerated without destabilising the system.

## **POLYMERS**

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Compositions of our invention may contain various polymers. In particular it is possible to incorporate useful amounts of polyelectrolytes such as uncapped polyacrylates or polymaleates. Such polymers may be useful because they tend to lower viscosity and because they have a detergent building effect and may have

anticorrosive or antiscaling activity. Unfortunately they also tend to break surfactant structure and cannot normally be included in structured surfactants in significant amounts without destabilising the system. We have discovered that relatively high levels of polyelectrolytes can be added to structured detergents in conjunction with deflocculated polymers and auxiliary stabilisers of the invention without destabilising the structure. This can provide stable products of even lower viscosity than can be achieved with the deflocculant and auxiliary stabiliser alone.

Some examples of polymers which may be included in the formulation are antiredeposition agents such as sodium carboxymethyl cellulose, antifoams such as silicone antifoams, enzyme stabilisers such as polyvinyl alcohols and polyvinyl pyrrolidone, dispersants such as lignin sulphonates and encapsulents such as gums and resins. We have found that milling aids such as sodium dimethylnaphthalene sulphonate/formaldehyde condensates are useful where the solid suspended in the composition requires milling as in the case of dye or pesticide formulations.

The amount of polymer added depends on the purpose for which it is used. In some cases it may be as little as 0.01% by weight, or even lower. More usually it is in the range of 0.1 to 10%, especially 0.2 to 5%, e.g. 0.5 to 2% by weight.

#### **OTHER DETERGENT ADDITIVES**

The solid suspending detergent compositions of our invention may comprise conventional detergent additives such as antiredeposition agents (typically sodium carboxymethyl cellulose), optical brighteners, sequestrants, antifoams, enzymes, enzyme stabilisers, preservatives, dyes, pigments, perfumes, fabric conditions, e.g. cationic fabric softeners or bentonite, opacifiers, bleach activators and/or chemically compatible bleaches. We have found that peroxygen bleaches such as sodium perborate, especially bleaches that have been protect e.g. by encapsulation, are more stable to decomposition in formulations according to our invention than in conventional liquid-detergents. Generally all conventional detergent additives which are dispersible in the detergent composition as solid particles or liquid droplets, in

excess of their solubility in the detergent, and which are not chemically reactive therewith may be suspended in the composition.

### **APPLICATIONS**

In addition to providing novel laundry detergents, fabric conditioners and scouring creams the stabilised structured surfactants of our invention may be used in toiletries, including shampoos, liquid soaps, creams, lotions, balms, ointments, antiseptics, dentifrices and styptics.

They provide valuable suspending media for dye and pigment concentrates and printing inks, pesticide concentrates and drilling muds. In the presence of dense dissolved electrolytes such as calcium bromide they are particularly useful for oilfield packing fluids (used to fill the gap between the pipe and the inside of the borehole, to protect the former from mechanical stresses) and completion fluids in oil well, or as cutting fluids or lubricants.

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The invention will be illustrated by the following examples.

### EXAMPLE 1

		%a.i.
"ACUSOL" <sup>®</sup> 842	non cross-linked acrylic acid/ethylacrylate copolymer	0.21
NaOH	50% solution	0.12
"LAUNDROSIL" <sup>®</sup> DGA	bentonite clay	1.67
"EMPIGEN" <sup>®</sup> BB	C <sub>12-14</sub> alkyl betaine	2.25
"BEVALOID" <sup>®</sup> XB16/01H	C <sub>16</sub> alkylthiolpolyacrylate	0.10
"EMPILAN" <sup>®</sup> KBE3	C <sub>12-14</sub> alkyl 3 mole ethoxylate	4.42
"WACKER" <sup>®</sup> S131	silicone antifoam	0.13
"BRIQUEST" <sup>®</sup> 543/25S	phosphonate	0.42
"TINOPAL" <sup>®</sup> CBS/X	optical brightener	0.06
	sodium carboxymethyl cellulose	0.08
	calcium chloride	0.21
"ALCALASE" <sup>®</sup> DX	protease enzyme	0.08
"TERMAMYL" <sup>®</sup> 300L	amylase enzyme	0.08
"PROXEL" <sup>®</sup>	preservative	0.02
	boric acid	0.83
Non-bio 32	perfume	0.33
STP/1L	sodium tripolyphosphate	20.8
	water	Balance



The above formulation was mobile and stable after three months storage. In the absence of the "ACUSOL" copolymer the composition underwent slow separation of a clear bottom layer over several weeks. This separation could not be prevented by increasing the amount of bentonite, even up to double the amount. In the absence of the bentonite some separation occurred which could be prevented by increasing the amount of "ACUSOL" copolymer.

### EXAMPLE 2

		%a.i.
"LAPONITE" <sup>®</sup> RD	synthetic layer silicate clay	0.2
"ACUSOL" <sup>®</sup> 842	non cross linked acrylic acid/ethyl acrylate copolymer	0.3
NaOH	sodium hydroxide	0.17
"EMPIGEN" <sup>®</sup> BB	C <sub>12-14</sub> alkyl betaine	2.75
"BEVALOID" <sup>®</sup> XB16/01H	alkylthiol polyacrylate	0.16
"EMPILAN" <sup>®</sup> KBE3	C <sub>12-14</sub> alcohol 3EO ethoxylate	4.95
"WACKER" <sup>®</sup> S131	silicone antifoam	0.25
	sodium tripolyphosphate	21.9
"BRIQUEST" <sup>®</sup> 543/25S	amino phosphonate	0.27
"REPELOTEX" <sup>®</sup> QCJ	soil release polymer	0.3
	water	Balance
Viscosity 1000cps (Brookfield Spindle4 100 rpm)		

The composition was stable on storage. Without the "ACUSOL" copolymer and "LAPONITE" synthetic clay the composition underwent rapid sedimentation. In the absence of the "ACUSOL" copolymer the composition could only be stabilised by the use of uneconomically high proportions of the clay.

In the absence of the clay, substantially higher concentrations of the "ACUSOL" copolymer were required which were significantly less cost effective than the mixture.

**CLAIMS**

1. A structured surfactant composition capable of suspending solids which comprises surfactant, water and, if required, electrolyte in relative proportions adapted to form a flocculated, dispersed lamellar and/or spherulitic structured surfactant system, and, in addition, sufficient of a deflocculant to inhibit the flocculation of said system characterised in that said composition comprises an effective amount of an auxiliary stabiliser which is a substantially non-cross linked, water-soluble copolymer having a hydrophilic backbone and sufficient C<sub>2-6</sub> hydrophobic organic side chains to permit entanglement of the polymer chain.
2. A composition according to claim 1 wherein said auxiliary stabiliser is a copolymer of an unsaturated carboxylic or dicarboxylic acid having from 3 to 6 carbon atoms and a C<sub>1</sub> to <sub>5</sub> alkyl ester of such an acid.
3. A composition according to claim 2 wherein said acid is acrylic acid.
4. A composition according to either of claims 2 and 3 wherein said ester is an ethyl, propyl or butyl ester of said acid.
5. A composition according to any of claims 2 to 4 wherein the ratio of said acid to said ester is from 0.01 to 10.
6. A composition according to any foregoing claim wherein said auxiliary stabiliser is present in a amount of from 0.05 to 5% by weight of the composition.
7. A composition according to any foregoing claim which additionally contains from 0.01 to 10% by weight of the composition of a clay.

8. A composition according to claim 7 wherein said clay is a water dispersible synthetic layer silicate.
  9. An auxiliary stabiliser for deflocculated structured surfactants which comprised from 10 to 90% by weight of a copolymer having a hydrophilic backbone and C<sub>2</sub> to 6 hydrophobic side chains, with from 90 to 10% by weight of a natural or synthetic clay.
-

# INTERNATIONAL SEARCH REPORT

International Application No

PCT/GB 00/02448

## A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 C11D17/00 C11D3/37 C11D3/12

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 C11D

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

WPI Data, EPO-Internal, PAJ

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	WO 91 08281 A (UNILEVER ) 13 June 1991 (1991-06-13) claims 1,2,4; example I	1-9
A	WO 91 09108 A (UNILEVER) 27 June 1991 (1991-06-27) abstract; example 1	1-6,9
A	US 5 599 784 A (BAINBRIDGE PETER) 4 February 1997 (1997-02-04) claims 1,2; examples 1,2	1-6,9



Further documents are listed in the continuation of box C.



Patent family members are listed in annex.

\* Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

Date of the actual completion of the international search

9 October 2000

Date of mailing of the international search report

20/10/2000

Name and mailing address of the ISA

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# INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/GB 00/02448

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
WO 9108281 A	13-06-1991	AU 641954 B AU 6607890 A BR 9007893 A CA 2069863 A DE 69005644 D DE 69005644 T EP 0504150 A ES 2062561 T JP 5502901 T ZA 9009738 A	07-10-1993 26-06-1991 15-09-1992 05-06-1991 10-02-1994 11-05-1994 23-09-1992 16-12-1994 20-05-1993 26-08-1992
WO 9109108 A	27-06-1991	AU 652543 B AU 6649290 A BR 9007902 A DE 69008735 D DE 69008735 T EP 0504159 A ES 2052279 T JP 2823355 B JP 5502249 T ZA 9009843 A	01-09-1994 18-07-1991 15-09-1992 09-06-1994 25-08-1994 23-09-1992 01-07-1994 11-11-1998 22-04-1993 26-08-1992
US 5599784 A	04-02-1997	US 5489397 A	06-02-1996

## PATENT COOPERATION TREATY

PCT

## NOTIFICATION OF ELECTION

(PCT Rule 61.2)

From the INTERNATIONAL BUREAU

To:

Commissioner  
 US Department of Commerce  
 United States Patent and Trademark  
 Office, PCT  
 2011 South Clark Place Room  
 CP2/5C24  
 Arlington, VA 22202  
 ETATS-UNIS D'AMERIQUE  
 in its capacity as elected Office

<b>Date of mailing (day/month/year)</b> 09 February 2001 (09.02.01)	<b>Applicant's or agent's file reference</b> CDK 1693
<b>International application No.</b> PCT/GB00/02448	<b>Priority date (day/month/year)</b> 24 June 1999 (24.06.99)
<b>International filing date (day/month/year)</b> 22 June 2000 (22.06.00)	
<b>Applicant</b> HATCHMAN, Kevan	

1. The designated Office is hereby notified of its election made:

☒ in the demand filed with the International Preliminary Examining Authority on:  
 20 December 2000 (20.12.00)

☐ in a notice effecting later election filed with the International Bureau on:  
 \_\_\_\_\_

2. The election ☒ was

☐ was not

made before the expiration of 19 months from the priority date or, where Rule 32 applies, within the time limit under Rule 32.2(b).

The International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland Facsimile No.: (41-22) 740.14.35	Authorized officer Juan Cruz Telephone No.: (41-22) 338.83.38
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## PCT

## INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference <b>CDK 1693</b>	<b>FOR FURTHER ACTION</b> see Notification of Transmittal of International Search Report (Form PCT/ISA/220) as well as, where applicable, item 5 b low.	
International application No. <b>PCT/GB 00/ 02448</b>	International filing date (day/month/year) <b>22/06/2000</b>	(Earliest) Priority Date (day/month/year) <b>24/06/1999</b>
Applicant <b>RHODIA CONSUMER SPECIALTIES LIMITED et al.</b>		

This International Search Report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This International Search Report consists of a total of 2 sheets.



It is also accompanied by a copy of each prior art document cited in this report.

## 1. Basis of the report

- a. With regard to the language, the international search was carried out on the basis of the international application in the language in which it was filed, unless otherwise indicated under this item.



the international search was carried out on the basis of a translation of the international application furnished to this Authority (Rule 23.1(b)).

- b. With regard to any nucleotide and/or amino acid sequence disclosed in the international application, the international search was carried out on the basis of the sequence listing :



contained in the international application in written form.



filed together with the international application in computer readable form.



furnished subsequently to this Authority in written form.



furnished subsequently to this Authority in computer readable form.



the statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.



the statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished

2. ☐ Certain claims were found unsearchable (See Box I).

3. ☐ Unity of invention is lacking (see Box II).

4. With regard to the title,



the text is approved as submitted by the applicant.



the text has been established by this Authority to read as follows:

5. With regard to the abstract,



the text is approved as submitted by the applicant.



the text has been established, according to Rule 38.2(b), by this Authority as it appears in Box III. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority.

6. The figure of the drawing to be published with the abstract is Figure No.



as suggested by the applicant.



because the applicant failed to suggest a figure.



because this figure better characterizes the invention.



None of the figures.

## INTERNATIONAL SEARCH REPORT

International Application No

PCT/GB 00/02448

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WPI Data, EPO-Internal, PAJ

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A ✓	US 5 599 784 A (BAINBRIDGE PETER) 4 February 1997 (1997-02-04) claims 1,2; examples 1,2 -----	1-6,9



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\*&amp;\* document member of the same patent family

Date of the actual completion of the international search

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- Date of mailing of the international search report

20/10/2000

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Fax: (+31-70) 340-3016

Authorized officer

Saunders, T



# INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/GB 00/02448

Patent document cited in search report		Publication dat	Patent family member(s)	Publication dat
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			DE 69005644 D	10-02-1994
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			EP 0504150 A	23-09-1992
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			JP 5502249 T	22-04-1993
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US 5599784	A	04-02-1997	US 5489397 A	06-02-1996